

EIS characterization of unusual behaviors of lithiated graphite|electrolyte interface

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Abstract

The negative electrodes of lithium-ion batteries are generally consisted of graphite, which intercalates lithium ions in the potential range [0, 0.3] V vs. Li/Li^+ . In order to better understand the phenomena linked with the intercalation, Electrochemical Impedance spectroscopy (EIS) is a widespread-used technic. It especially allows to study the graphite composite electrode|electrolyte interface evolution versus the potential electrode.

The present paper deals with the characterization of a graphite composite electrode in a newly designed three-electrode button-type cell. In order to follow the evolution of the graphite|electrolyte interface, several spectra have been plotted during the two first reductions and oxydations of the electrochemical system.

Typical impedance spectra (figure 1) exhibit a high-frequency (HF) impedance loop, which doesn't evolve much over the first reduction. A middle-frequency (MF) loop appears as the cathodic polarization of the carbon electrode increases, whom characteristics are potential dependent. In the lower frequency (LF) region, a near vertical straight line is observed. During the first oxydation, that is the first de-intercalation of lithium ions out of graphite, the HF loop decreases dramatically when [0.05, 0.1] V vs. Li/Li^+ is reached (figure 2). On the other hand, the next reduction shows a large increase of the HF loop in the same potential range. This range corresponds to the phase transition between $\text{Li}_{0.5}\text{C}_6$ and LiC_6 .

Other spectra have been drawn using the same electrochemical system, the electrolyte excepted, which has been contaminated with some water ppm (respectively 100, 500 and 1000) for the experiment. The results are original : an inductive loop appears in the MF region whatever the added water content may be (figure 3).

All these points are discussed in the present paper.

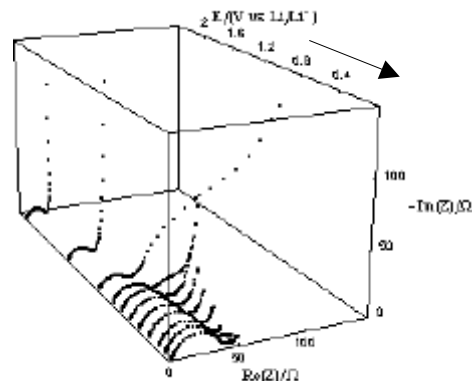


Fig. 1. Impedance spectra of the three-electrode cell at different potentials during the first intercalation of lithium ions into carbon.

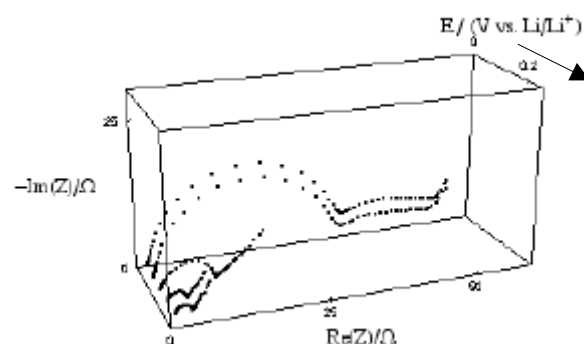


Fig. 2. Impedance spectra of the three-electrode cell at different potentials during the first de-intercalation of lithium ions out of carbon.

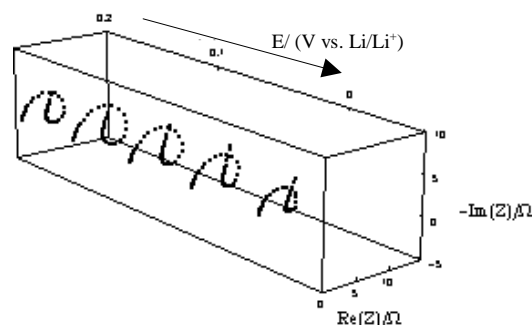


Fig. 3. Impedance spectra of the three-electrode cell at different potentials during the first intercalation of lithium ions into carbon. 500 water ppm have been added in the electrolyte used here.